Indian J. Phys. **67A** (2), 119-123 (1993)

A study on local moments and charge fluctuation in mixed-valence systems

N K Ghosh and R L Sarkar
Department of Physics, University of Kalyani, Kalyani-741 235, West Bengal, India

Received 24 September 1992, accepted 2 November 1992

Abstract : An attempt is made to study local moments and charge fluctuation in mixed-valence systems. Model Hamiltonian approach has been accepted. Role of *f-f* Coulomb interaction and electron-phonon (EP) interaction is investigated. A comparison is also made with few existing results.

Keywords : Mixed-valence systems, local moments and charge fluctuation, Coulomb interaction, electron-phonon interaction

PACS Nos. : 71.70.ch, 74.40 +k

1. Introduction

Plenty of studies on mixed-valence (MV) phenomena exhibited by rare-earth intermetallics and compounds have been done using various model Hamiltonians [1-3]. Recently, these model Hamiltonians have been successfully used to explain the characteristic features of local moments and charge fluctuations which are observed in the majority of rare-earth materials [4-6].

Blankenbecler *et al* [7] considered a one dimensional, symmetric, periodic Anderson model and studied the ground state properties using stochastic Monte-Carlo techniques. They have shown that the ground state exhibits short-range magnetic correlations and that the local *f*-electron spin moments are compensated by correlations with other *f*-electrons as well as band electrons leading to a nonmagnetic ground state. NIE and WEI [8] used the local approach to study the correlation effects of the periodic Anderson lattice. Their results showed that the Coulomb interaction U leads to a formation of local moments and a reduction of charge fluctuation. It is in agreement with Yamada and Yosida [9] who investigated the ground-state properties of the periodic Anderson Hamiltonian using higher-order perturbation theory.

In the present work, the possible nature of local moments and charge fluctuation in MV systems has been investigated. We have considered a two-site model Hamiltonian [10], which incorporates both the Falicov-Kimball term and the electron-phonon (EP) interaction term. A suitable representative state is chosen [10] for which the exact calculation is

admissible. Effect of different interactions on local moments and charge fluctuation have been studied.

2. Formulation

We have considered the following model Hamiltonian :

$$H = H_0 + H_v \tag{1}$$

where $$H_0 = \sum_{i,\sigma}(E_0 d^\dagger_{i\sigma} d_{i\sigma} + E f^\dagger_{i\sigma} f_{i\sigma}) + G \sum_{i,\sigma,\sigma'} d^\dagger_{i\sigma} d_{i\sigma} f^\dagger_{i\sigma'} f_{i\sigma'}$$

$$+ U_{ff} \sum_i f^\dagger_{i\uparrow} f_{i\uparrow} f^\dagger_{i\downarrow} f_{i\downarrow} + W \sum_{\substack{i,j,\sigma \\ i\neq j}} d^\dagger_{i\sigma} d_{j\sigma} \tag{2}$$

and, $$H_v = V \sum_{\substack{i,j,\sigma \\ i\neq j}} (f^\dagger_{i\sigma} d_{j\sigma} + d^\dagger_{j\sigma} f_{i\sigma}) - g \sum_{\substack{i,j,\sigma,\sigma' \\ i\neq j}} n^d_{i\sigma'} (f^\dagger_{i\sigma} d_{j\sigma} + d^\dagger_{j\sigma} f_{i\sigma}) . \tag{3}$$

E_0, E are the d-level and f-level energy, respectively, G is the strength of the f-d Coulomb interaction, U_{ff} the Coulomb repulsion between the f-electrons of opposite spins on the same site, W the strength of the hopping interaction between d-electrons at different sites. V the f-d hybridisation parameter, and g is the electron-phonon interaction parameter. We have restricted ourselves to the case $<n^f_i> + <n^d_i> = 1$. Also, throughout our calculation $i, j = 1, 2$ and $\sigma, \sigma' = \uparrow, \downarrow$.

The representative two-site spin state is taken in the form

$$| n^f_{1\uparrow} n^f_{1\downarrow} n^d_{1\uparrow} n^d_{1\downarrow} n^f_{2\uparrow} n^f_{2\downarrow} n^d_{2\uparrow} n^d_{2\downarrow} > \tag{4}$$

and the eigenvectors of H are represented as a linear combinations of vectors (4).

In such a way we have solved the eigen equation

$$H | E_\alpha > = E_\alpha | E_\alpha >, \alpha = 1, \ldots\ldots, 28. \tag{5}$$

The ground state $|\psi>$ is constructed for which E_α has the lowest value.

The local moments S_m and the charge fluctuation Q_f are calculated using the following formulas,

$$S_m = 3/4\ (<n^f_i> - 2 <n^f_{i\uparrow} n^f_{i\downarrow}>),$$

$$Q_f = <n^f_i> (<n^f_i> - 1) + 2 <n^f_{i\uparrow} n^f_{i\downarrow}>.$$

3. Results and discussions

Figure 1 shows the variation of the local moments S_m with U_{ff} for different hybridisation strengths. The local moment is increased with increasing U_{ff} for fixed V. The Coulomb interaction U_{ff} reduces the probability of finding two f-electrons with opposite spins at the same site, and leads to a formation of local moments on the f-orbitals. Since we are considering the system in the region of phase transition, the hybridisation V has two

opposing effects on f-electron occupation number in these two phases (insulating and metallic). As the value of U_{ff} increases, the system gradually enters into the metallic phase. $<n_i^f>$ as well as $<n^f_{i\uparrow}\, n^f_{i\downarrow}>$ decreases with V in the insulating phase and increases with V in the metallic phase. Finally, we obtain the characteristics of S_m as shown in the figure.

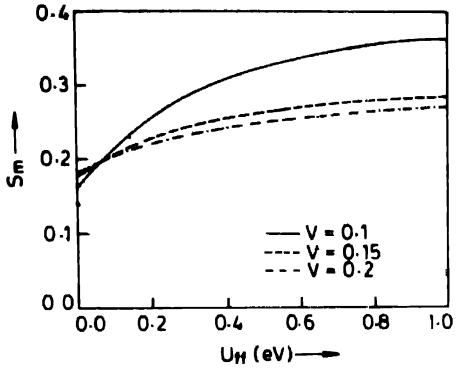

Figure 1. S_m as a function of U_{ff} for $T = 0$ K and for various values of V (in eV). $E_0 = 0$ eV, E = –0.1 eV, $G = 0.5$ eV, $W = 0.06$ eV, $g = 0$ eV

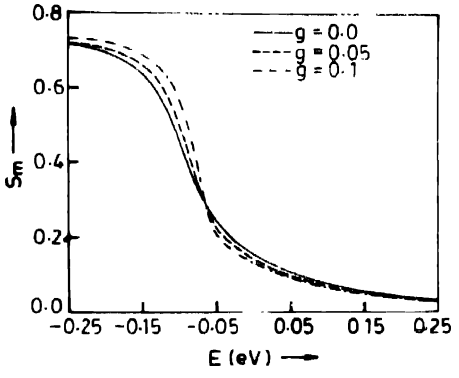

Figure 2. S_m as a function of E for $T = 0$ K and for various values of g (in eV). $E_0 = 0$ eV, $G = 0.25$ eV, $W = 0.06$ eV, $U_{ff} = 1.0$ eV, $V = 0.1$ eV.

S_m varies with E in the same way as $<n_i^f>$ as is apparent in Figure 2, where local moment has been plotted against E for different EP interaction. As the value of E increases, $<n_i^f>$ decreases which reduces the formation of local moments on the f-orbitals. The fluctuation of $<n^f_{i\uparrow}\, n_{f\downarrow}>$ with E, and EP interaction is very small compared to $<n_i^f>$ and so in the present case, $<n_i^f>$ determines S_m. In the insulating phase, EP interaction increases S_m following $<n_i^f>$. With the increase of E phase transition occurs and S_m decreases with g.

In Figure 3, we have plotted the charge fluctuation Q_f vs U_{ff} for different hybridisation strengths. The result shows that the charge fluctuation Q_f is decreased with increasing U_{ff} for fixed V. As the value of U_{ff} increases, $<n_i^f>$ as well as $<n_i^{f}{}_{\uparrow}\, n_i^{f}{}_{\downarrow}>$

decreases. These make Q_f decrease with U_{ff}. The result is in agreement with local approach method in periodic Anderson lattice [8]. Here also, in the insulating phase, Q_f decreases with V and in the metallic phase, Q_f increases with V.

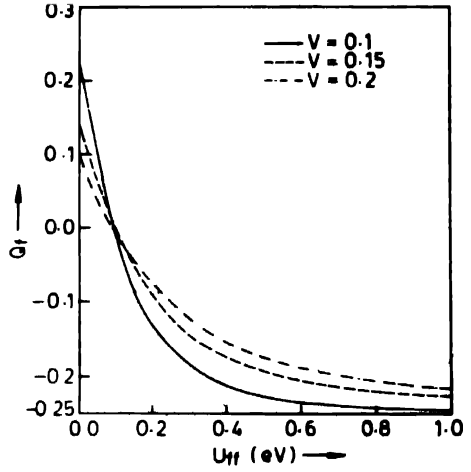

Figure 3. Q_f as a function of U_{ff} for $T = 0$ K and for various values of V (in eV). E_0 0 eV, $E = -0.1$ eV, $G = 0.5$ eV, $W = 0.06$ eV, $g = 0$ eV.

Curves of the charge fluctuation against E are presented in Figure 4 from which we find that with the increase of E, Q_f decreases from a value close to zero and after a certain E, it increases towards the value zero. Here, as charge fluctuation is primarily determined by $<n_i^f>$, so, in the insulating or metallic phase the charge fluctuation should approach zero. For large negative E, EP interaction keeps the value of $<n_i^f>$ close to unity, and after the transition keeps a value of $<n_i^f>$ close to zero. In both the cases Q_f should be closer to zero than that for smaller EP interaction as is apparent in Figure 4.

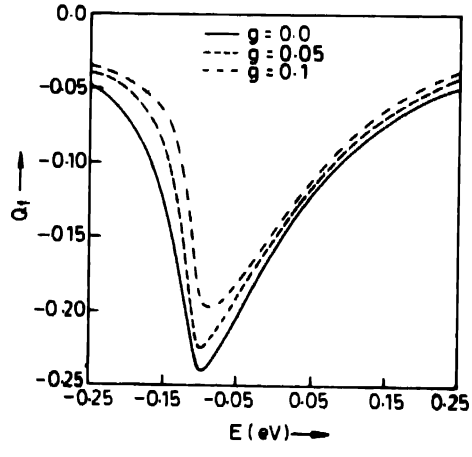

Figure 4. Q_f as a function of E for $T = 0$ K and for various values of g (in eV). $E_0 = 0$ eV, $G = 0.25$ eV, $W = 0.06$ eV, $U_{ff} = 1.0$ eV, $V = 0.1$ eV.

Acknowledgment

We are thankful to the University Grants Commission, New Delhi, for financial help.

References

[1] I Singh, A K Ahuja and S K Joshi 1980 *Solid State Commun.* **34** 65

[2] A P G Kutty 1984 *J. Phys. Chem. Solids* **45** 121

[3] N K Ghosh, S C Ghosh and R L Sarkar 1992 *Phys. Stat. Sol. (b)* **71** 107

[4] F Steglich, J Aarts, C D Bredl, W Lieke, D Meschede, W Franz and H Schafer 1979 *Phys. Rev. Lett.* **43** 1892

[5] H R Ott, H Rudigier, P Delsing and Z Fisk 1984 *Phys. Rev. Lett.* **52** 1551

[6] G R Stewart, Z Fisk, J O Wills and J L Smith 1984 *Phys. Rev. Lett.* **52** 679

[7] R Blankenbecler, J R Fulco, W Gill and D J Scalapino 1987 *Phys. Rev. Lett.* **58** 411

[8] Hui-Quan NIE and Guo-Zhu WEI 1989 *J. Magn. Magn. Mater.* **78** 415

[9] K Yamada and K Yosida 1981 *Proceedings of the Third Symposium, Mount Fuji, Japan, 1980* ed. T Moriya (Berlin : Springer-Verlag) p 210

[10] S C Ghosh , N K Ghosh and R L Sarkar 1990 *Phys Stat. Sol (b)* **161** 661